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DISCUSSION OF THE AMENDMENT

Claim 57 has been amended by deleting "wherein the binder resin comprises units from a monomer having a Brönsted acidic group or a Brönsted basic group" and inserting that the binder resin is --obtained by addition polymerization--. While the term --addition polymerization-- does not appear *per se*, it is notoriously well-known from the types of monomers and the polymerizations described, such as emulsion polymerization, that resins prepared therein are by addition polymerization. See Morrison and Boyd, *Organic Chemistry*, Allyn and Bacon, Inc., Boston (1959), pp. 184-191 (copy enclosed).

Claim 27 has been amended, and Claim 57 has been further amended, by replacing a period with a semi-colon, as required by the Examiner.

New Claims 68-69 have been added. Claim 68 is supported by Claim 27 and the binder limitations of Claim 1, as supported in the specification at page 41, lines 5-8. Claim 69 is supported by original Claim 16 and the specification at page 14, line 20 ff.

No new matter has been added by the above amendment. Claims 1-43 and 45-69 will be pending. Claims 27-30, 32, 34, 37, 38, 40, 42 and 57-69 will be active; Claims 1-26, 31, 33, 35, 36, 39, 41, 43, and 45-56 stand withdrawn from consideration.

REMARKS

As recited in Claim 57, an embodiment of the presently-pending invention is a toner comprising: a binder resin obtained by addition polymerization, and a particulate wax, wherein the toner has a volume-average particle diameter of from 3 to 12 μm , and a half value width of a number-average particle diameter of particulate wax contained therein, when a cross section of the toner is observed, of 0.06 μm or less, and wherein a distribution of particulate wax having an average particle diameter of 0.01 μm or more throughout the toner satisfies the following equation:

$$(A/B)/(C/D) \leq 0.1$$

wherein A is total area of particulate wax contained in an outermost layer of the toner to a depth of 0.1 μm ;

B is total area of said outermost layer of the toner;

C is total area of particulate wax contained in a remainder of the toner (at a depth of greater than 0.1 μm from the surface of the toner); and

D is total area of said remainder of the toner,

wherein all areas are measured as observed in a cross section of said toner through a center point of said toner.

In effect, the toner is substantially free of wax particles at its outermost part, specifically in the area of the depth of 0.1 μm from the surface of the toner, as described in the specification at page 47, line 3, through page 48, line 7, and Fig. 4.

The rejections of Claims 57-62 and 65 under 35 U.S.C. § 102(e) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious over, U.S. 2002/0028402 A1 (Matsuoka et al); and under 35 U.S.C. § 103(a), of Claim 63 as unpatentable over Matsuoka et al combined with U.S. 5,213,932 (Shimazaki); of Claim 64 as unpatentable over Matsuoka

et al combined with JP 59-165069 (JP '069); and of Claim 66 as unpatentable over Matsuoka et al combined with U.S. 5,547,802 (Kawase et al), are all respectfully traversed.

Matsuoka et al disclose a toner containing a releasing agent, which is preferably a wax, and which has a content existing at the surface of the toner particles of 0 to 30% by weight, preferably from 1% to 10% by weight ([0058]). Matsuoka et al define the surface of the toner particles to mean a layer extending from the top to a depth of 0.1 μm ([0059]). The Examiner particularly relies on the exemplified yellow toner in Table 2, wherein the amount of releasing agent on the surface of the toner particles is 4.2% by weight and the releasing agent dispersion diameter is 0.8 μm . The Examiner finds that this exemplified yellow toner inherently meets the terms of the present claims.

Shimazaki is relied on for its disclosure of a magenta colorant. JP '069 is relied on for its disclosure of a magenta colorant. Kawase et al is relied on for a disclosure of ratio of volume mean particle diameter to number average particle diameter.

The Examiner concedes that Matsuoka et al does not disclose the presently-recited "half value width" limitation, i.e., a half value width of a number-average particle diameter of particulate wax contained therein, when a cross section of the toner is observed, of 0.06 μm or less, but presumes that Matsuoka et al's toner meets this limitation. Applicants respectfully submit that the "half value width" of Matsuoka et al is much larger than 0.06 μm , based on the following reasons:

i) Fig. 2 in Matsuoka et al shows a distribution of the dispersion diameter (d_1) of the releasing agent particles that is relatively wide.

ii) Matsuoka et al discloses "... the releasing agent is dispersed with a particle diameter of preferably 3 μm or less and more preferably in a range from 0.1 to 2 μm " ([0060]). Matsuoka et al's measuring method differs from that of the present invention regarding wax diameter. The method of the present invention is determined by observation

of the cross section of the toner, as recited in Claim 57; on the other hand, Matsuoka et al's method is as disclosed in ([0054]) therein. Despite differences in measuring method, the range of 0.1 to 2 μm of Matsuoka et al is much wider than the 0.06 μm maximum recited in Claim 57.

iii) The Examiner refers to page 49, lines 4-11 of the specification. However, immediately thereafter, Applicants describe that where a particulate wax is co-agglomerated with primary polymer particles, the distribution of the number-average particle diameter of the particle wax becomes wider.

See also Reference Synthesis Example, beginning at page 125, line 2, of the specification. In the Reference Synthesis Example and Fig. 8, the number-average particle diameter of particulate wax observed was 201 nm, which was relatively small compared to average particle diameter of the same in the wax dispersion 15, and a half value width of the number-average particulate diameter in the toner was 100 nm, or 0.1 μm .

With such a small half value width of a number average particle diameter of particulate wax, as recited in Claim 57, the toner of the present invention has excellent releasability, excellent blocking resistance and the apparatus is hardly polluted, as described at page 50, lines 15 to 22, of the specification.

While Matsuoka et al may disclose that the number of particles of releasing agent in the surface layer up to 0.1 μm is much less than the number of particles at a depth of 0.1 μm or more, there is no reason to believe that the above-discussed ratio is satisfied. Nor does Matsuoka et al disclose a method of producing a toner having a releasing agent content in the outermost layer of the toner, less than that of the remainder.

In addition to all the above, it is also noted that Matsuoka et al's toner includes a binder resin which contains a linear resin and a non-linear resin; while styrene resin, acrylic resin, styrene/acryl-type resin, are listed as applicable resins, polyester resins are described as

preferable ([0031]), and all of the examples are limited to polyester resins as binder resins. It is well-known that polyesters are condensation polymers, not addition polymers. Nor does Matsuoka et al disclose their binder resin as comprising an agglomerate of particles comprising at least primary polymer particles, and at least one layer of a particulate resin coated on a substantial surface portion of said agglomerate of particles, or wherein the primary polymer particles comprise units from a monomer having a Brönsted acidic group or a Brönsted basic group.

For all the above reasons, it is respectfully requested that the rejections over Matsuoka et al alone, or combined with other prior art be withdrawn.

The rejection of Claims 57-67 under 35 U.S.C. § 112, first paragraph, as failing to satisfy the description requirement therein, is respectfully traversed. Indeed, the rejection is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that it be withdrawn.

The objection to Claims 27 (and claims dependent thereon) and 57 (and claims dependent thereon), is respectfully traversed. Indeed, the objection is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that it be withdrawn.

Applicants gratefully acknowledge the Examiner's indication of allowability of the subject matter of Claims 27-30, 32, 34, 37, 38, 40 and 42. Nevertheless, Applicants respectfully submit that all of the presently pending and active claims in this application are

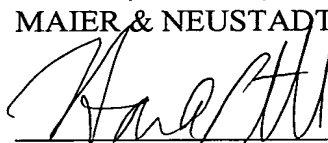
Application No. 09/737,579

Reply to Office Action of November 18, 2003

now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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ORGANIC CHEMISTRY

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6.19 Free radical addition to conjugated dienes: reactivity

If BrCCl_3 is allowed to react with a 50:50 mixture of 1,3-butadiene and a simple alkene like 1-octene, addition occurs almost exclusively to the 1,3-butadiene. Evidently the $\cdot\text{CCl}_3$ radical adds much more rapidly to the conjugated diene than to the simple alkene. Similar results have been observed in a great many radical additions.

How can we account for the unusual reactivity of conjugated dienes? In our discussion of halogenation of the simple alkanes (Sec. 3.26), we found that not only orientation but also relative reactivity was related to the stability of the free radical formed in the first step. On this basis alone, we might expect addition to a conjugated diene, which yields a stable allyl free radical, to occur faster than addition to a simple alkene.

On the other hand, we have just seen (Sec. 6.15) that conjugated dienes are more stable than simple alkenes. On this basis alone, we might expect addition to conjugated dienes to occur more slowly than to simple alkenes.

The relative rates of the two reactions depend chiefly upon the E_{act} 's. Stabilization of the incipient allyl free radical lowers the energy level of the transition state; stabilization of the diene lowers the energy of the reactants. Whether the net E_{act} is larger or smaller than for addition to a simple alkene depends upon which is stabilized more (see Figure 6.7).

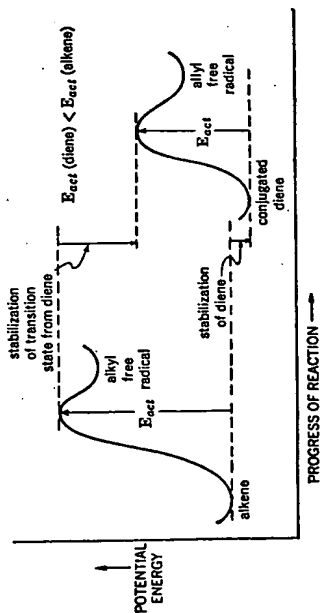


FIGURE 6.7. Molecular structure and rate of reaction. Stability of transition state parallels stability of free radical: more stable free radical formed faster.

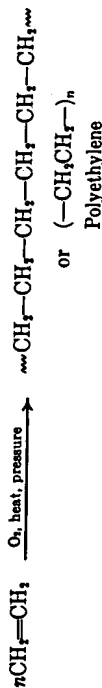
The fact is that conjugated dienes are more reactive than simple alkenes. In the present case, then — and in most cases involving alkenes and free radicals, or alkenes and carbonium ions — the factors stabilizing the transition state are more important than the factors stabilizing the reactant. However, this is *not always* true.

POLYMERIZATION

6.20 Free radical polymerization of alkenes

One of the most important reactions of simple alkenes and conjugated dienes is polymerization.

When ethylene is heated under pressure with oxygen, there is obtained a compound of high molecular weight (about 20,000), which is essentially an alkane with a very long chain. This compound is made up of many

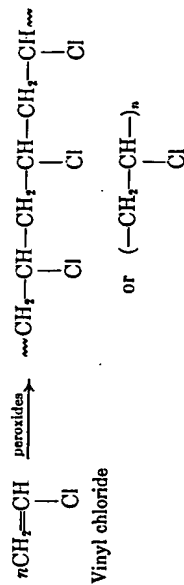


ethylene units and hence is called *polyethylene* (poly = many). As Polythene it is familiar to most of us as the plastic material of flexible bottles, children's toys, and packaging films.

The formation of polyethylene is a simple example of the process called **polymerization**: the joining together of many small molecules to make very large molecules. The compound composed of these very large molecules is called a **polymer** (from the Greek, *poly* + *meros*, many parts). The simple compounds from which polymers are made are called **monomers** (*mono* = one).

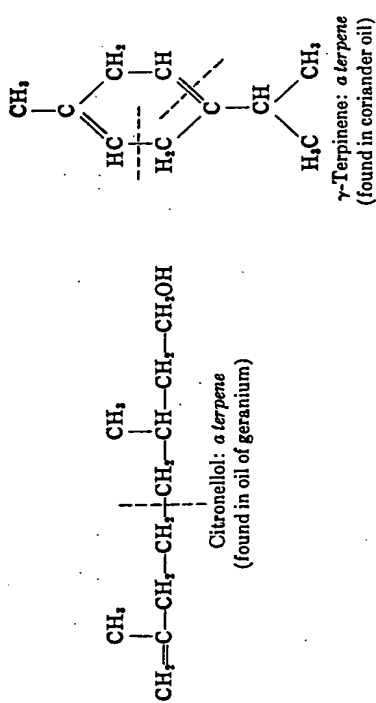
The particular kind of polymerization undergone by ethylene, in which many molecules of monomer are simply added together, is called **addition polymerization**. (Later on, in Sec. 25.5, we shall encounter **condensation polymerization**, in which monomer molecules combine with the loss of some simple molecules, usually water.) Addition polymerization is one of the most important reactions of alkenes and conjugated dienes alike — and, indeed, of compounds of all kinds that contain carbon-carbon double bonds.

Polymerization of substituted ethylenes yields compounds whose structures contain the long chain of polyethylene, with substituents attached at more or less regular intervals. For example, vinyl chloride



yields *polyvinyl chloride*, used in phonograph records, enamels, and as the basis of Koroseal (shower curtains, raincoats, and a substitute for rubber). Many other groups (e.g., $-\text{COOCH}_3$, $-\text{CN}$, $-\text{C}_6\text{H}_5$) may be attached to the doubly-bonded carbons. These substituted ethylenes polymerize more or less readily, and yield plastics of widely differing physical properties and uses, but the polymerization process and the structure of the polymer are basically the same as for ethylene or vinyl chloride.

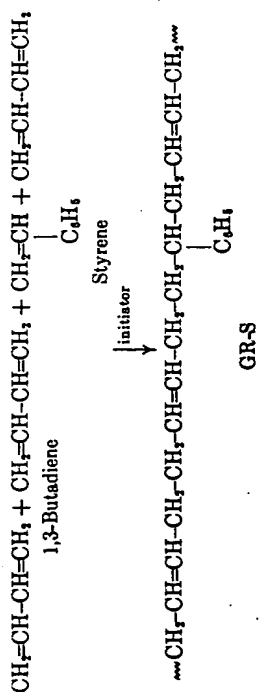
Polymerization requires the presence of a small amount of an initiator. Among the commonest of these initiators are peroxides, the same compounds that bring about free radical addition to alkenes (Sec. 5.7). Here, too, a peroxide is believed to function by breaking down to form a free



6.22 Copolymerization

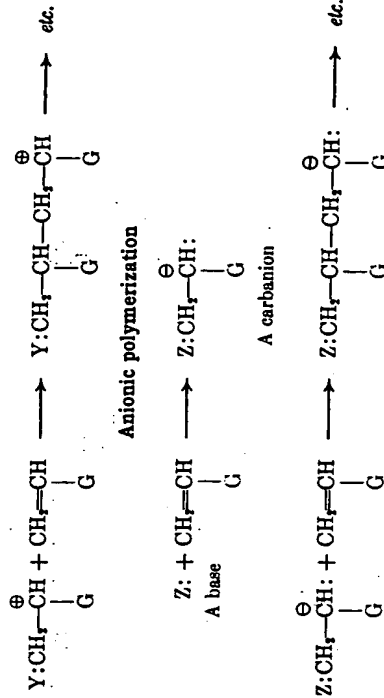
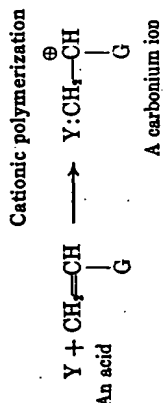
A further way to modify the properties of a polymer is through the process of copolymerization. Here two (or more) unsaturated compounds are mixed and allowed to polymerize together. The polymer formed contains units of both kinds, alternating more or less regularly along the chain.

A particularly important copolymer, for example, is one between butadiene and *styrene* ($\text{C}_6\text{H}_5-\text{CH}=\text{CH}_2$). This material, which is generally composed of about three parts of butadiene and one part of styrene, is GR-S, during World War II the most important of the synthetic rubbers developed to replace unavailable natural rubber. Since the war the synthetic rubber industry has continued to expand, largely because of the demands of the automobile industry.

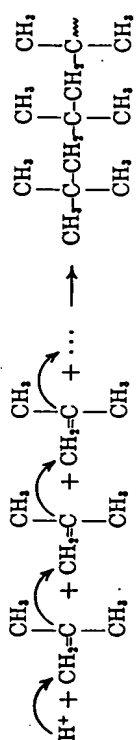


6.23 Ionic polymerization

So far, we have discussed only the kind of addition polymerization that takes place by way of free radicals. By use of different initiators or catalysts, however, it is possible to bring about polymerization that involves intermediate ions: either positive ions (*cations*), when the catalyst is an acid; or negative ions (*anions*), when the catalyst is a base.

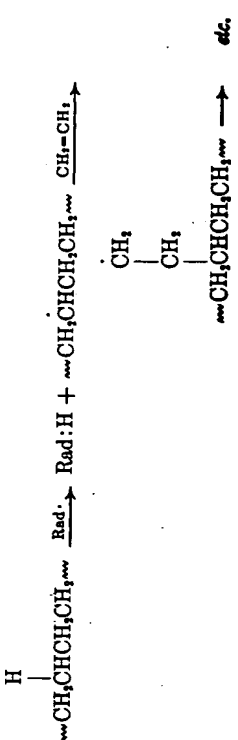


Until 1953, almost all addition polymerizations of commercial importance were of the free radical type. A notable exception was the acid-catalyzed (cationic) polymerization of isobutylene with a little butadiene to yield *butyl rubber*, used to make automobile inner tubes. We recognize this process as an extension of the dimerization we discussed earlier (Sec. 5.17).



Since 1953, however, ionic polymerization has grown to such an extent that it has revolutionized the field of polymerization. Following discoveries by Karl Ziegler (of the Max Planck Institute for Coal Research) and by Giulio Natta (of the Polytechnic Institute of Milan), catalysts have been developed that permit control of the polymerization process to a degree never before possible. These catalysts include such substances as a triethylaluminum-titanium trichloride complex and finely divided lithium metal. Let us look briefly at just two aspects of these new developments, which illustrate the importance of the basic concepts of structural theory.

First, there is the matter of *chain branching*. Polyethylene made by the free radical process has highly branched chains. At the high temperature required for this particular polymerization, the growing free radicals not only *add* to the double bond of a monomer molecule but also *abstract* hydrogen from a chain already formed.



This abstraction generates a free radical center from which a branch can now grow. These highly branched polyethylene molecules fit together poorly and in a random way; the compound is said to have low *crystallinity*. It has a low melting point, and is mechanically weak.

In contrast, the newer polyethylene — made via ions instead of free radicals, and under mild conditions — is unbranched. It is highly crystalline, has a higher melting point, and is mechanically stronger.

A second, far-reaching development in the new ionic polymerization is *stereochemical control*. Propylene, for example, could polymerize to any of three different arrangements (Figure 6.8): *isotactic*, with all methyl groups on one side of an extended chain; *syndiotactic*, with methyl groups alternating regularly from side to side; and *atactic*, with methyl groups distributed at random.

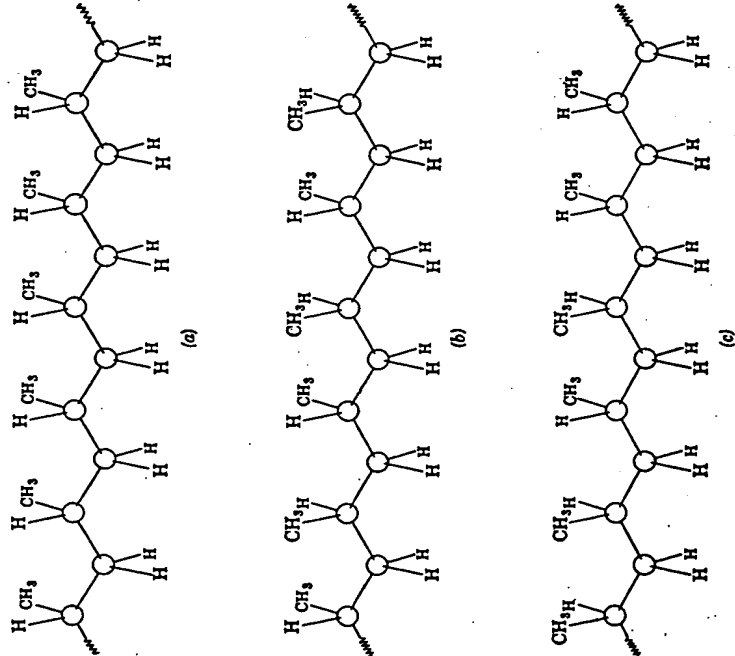
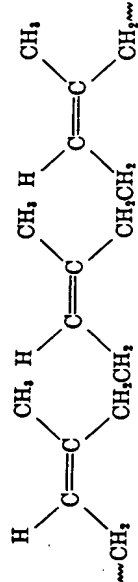


FIGURE 6.8. Polypropylene. (a) Isotactic. (b) Syndiotactic. (c) Atactic.

By use of catalysts whose molecules are mounted on a crystalline solid, isotactic polymers can be made. By use of catalysts mounted on an amorphous solid, atactic polymers are formed. Isotactic polypropylene is a highly crystalline, high-melting material that forms strong fibers. Atactic polypropylene is a soft, elastic, rubbery material.

For the first time, isoprene has been polymerized to a material virtually identical with natural rubber: *cis*-1,4-polyisoprene.

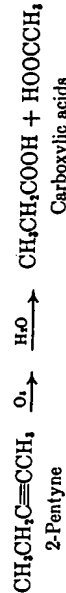


"A chemist setting out to build a giant molecule is in the same position as an architect designing a building. He has a number of building blocks of certain shapes and sizes, and his task is to put them together in a structure to serve a particular purpose. . . . What makes high polymer chemistry still more exciting just now is that almost overnight, within the last few years, there have come discoveries of new ways to put the building blocks together — discoveries which promise a great harvest of materials that have never existed on the earth." (Giulio Natta, *Scientific American*, September 1957, p. 98.)

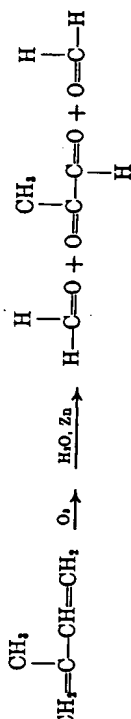
5.2.4 Analysis of alkynes and dienes

Alkynes and dienes respond to characterization tests in the same way as alkenes: they decolorize bromine in carbon tetrachloride without evolution of hydrogen bromide, and they decolorize cold, neutral, dilute potassium permanganate. They are, however, more unsaturated than alkenes. This property can be detected by determination of their molecular formulas (C_nH_{2n-2}) and by a quantitative hydrogenation (two moles of hydrogen are taken up per mole of hydrocarbon).

Proof of structure is best accomplished by the same degradative methods that are used in studying alkenes. Upon ozonolysis alkynes yield carboxylic acids, whereas alkenes yield aldehydes and ketones. For example:



Ozonolysis of dienes yields aldehydes and ketones, including double-ended ones containing two C=O groups per molecule. For example:



A terminal alkyne ($\text{RC}\equiv\text{CH}$) is characterized, and differentiated from isomers, by its conversion into insoluble silver and cuprous acetylides (Sec. 6.10).

Problem 6.15 Contrast the ozonolysis products of the following isomers: (a) 1-pentyne; (b) 2-pentyne; (c) 3-methyl-1-butyne; (d) 1,3-pentadiene; (e) 1,4-pentadiene; (f) isoprene (2-methyl-1,3-butadiene).

Problem 6.16 Predict the ozonolysis products from polybutadiene, $(C_4H_6)_n$: (a) if 1,2-addition is involved in the polymerization; (b) if 1,4-addition is involved.

Problem 6.17 Ozonolysis of natural rubber yields chiefly (90%) the compound shown on the next page. What does this tell us about the structure of rubber?